Wide-voltage-window reversible control of electronic transport in electrolyte-gated epitaxial BaSnO$_3$

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I. INTRODUCTION

The perovskite oxide semiconductor BaSnO$_3$ (BSO) has attracted substantial interest since the 2012 discovery of outstanding room temperature electron mobility [1–3]. The mobility ($\mu$) reaches 320 cm$^2$ V$^{-1}$ s$^{-1}$ at a Hall electron density $n \approx 10^{20}$ cm$^{-3}$ in La-doped bulk single crystals, the largest room temperature value among perovskite oxides [1]. Retention of high $\mu$ at this very high $n$ also generates exceptional conductivity [4]. The outstanding mobility is thought to derive from relatively small electron effective mass (recently reported to be $\sim 0.2 m_e$, where $m_e$ is the free electron mass [5,6]) in the dispersive Sn $5s$-derived conduction band [7–9], as well as low electron-phonon scattering rate [4,8]. The high mobility, wide band gap ($\sim 3$ eV), and high optical transparency [2] render BSO a candidate for several applications, including high mobility transistors, transparent electronics, power electronics, and as a component in complex oxide heterostructures. Recently, $n$ doping of bulk crystals has also been demonstrated with rare-earth dopants (Nd and Pr, which are magnetic) and oxygen vacancies (V$_O$), going beyond La [10].

The wide gap perovskite semiconductor BaSnO$_3$ has attracted much interest since the discovery of room temperature electron mobility up to 320 cm$^2$ V$^{-1}$ s$^{-1}$ in bulk crystals. Motivated by applications in oxide heterostructures, rapid progress has been made with BaSnO$_3$ films, although questions remain regarding transport mechanisms and mobility optimization. Here we report on a detailed study of epitaxial BaSnO$_3$ electric double layer transistors based on ion gel electrolytes, enabling wide-doping-range studies of transport in single films. The work spans an order of magnitude in initial $n$ doping ($\sim 10^{19}$ up to $10^{20}$ cm$^{-3}$, with both oxygen vacancies and La), film thicknesses from 10–50 nm, and measurements of resistance, Hall effect, mobilities, and magnetoresistance. In contrast with many oxides, electrolyte gating of BaSnO$_3$ is found to be essentially reversible over an exceptional gate voltage window (approaching $\pm 4$ V), even at 300 K, supported by negligible structural modification in operando synchrotron x-ray diffraction. We propose that this occurs due to a special situation in BaSnO$_3$, where electrochemical gating via oxygen vacancies is severely limited by their low diffusivity. Wide-range reversible modulation of transport is thus achieved (in both electron accumulation and depletion modes), spanning strongly localized, weakly localized, and metallic regimes. Two-channel conduction analysis is then combined with self-consistent Schrödinger-Poisson and Thomas-Fermi modeling to extract accumulation layer electron densities and mobilities. Electrostatic electron densities approaching $10^{14}$ cm$^{-2}$ are shown to increase room temperature mobility up to a factor of $\sim 24$. These results lay the groundwork for future studies of electron-density-dependent phenomena in high mobility BaSnO$_3$, and significantly elucidate oxide electrolyte gating mechanisms.

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their their n dependence, improving understanding of mobility limiting factors [4]. Systematic control over n in BSO is thus very useful, both to better understand mobility limits and to study doping-dependent phenomena.

Naturally given the above, BSO-based field-effect transistors (FETs) have been fabricated, seeking not only to explore device potential, but also to electrostatically control n in single films, independent of chemical doping. BSO-based FETs with A12O3, HfO2, LaInO3, SrTiO3, and parylene dielectrics have been reported, yielding ON/OFF ratios up to 107 [24–28], sub-threshold swing of 0.65 V dec−1 [27], and field-effect µ up to 90 cm2 V−1 s−1 [27]. While these are promising developments, from the perspective of fundamental studies of n-dependent transport such FETs are limited to n ∼ 1013 cm−2 (≈1019 cm−3) [26], modest compared to chemical doping limits in BSO. Other approaches are thus being explored, including ferroelectric gating of BSO (potentially increasing n) [29], and polarization doping in heterostructures such as LaInO3/ BaSnO3 [30,31].

A complementary approach, now established as highly effective in wide-range modulation of carrier density in oxides (and indeed more broadly), is electrolyte gating in electric double layer transistors (EDLTs) [32,33]. In this approach, the dielectric in a FET is replaced with an electrolyte, often an ionic liquid or ion gel. The latter are simply ionic liquids in polymer networks, generating easily processed solid electrolytes with high ionic mobility [33,34]. When a gate voltage (Vg) is applied in an oxide EDLT (positive, for example), cations in the ionic liquid or gel are driven to the surface of the oxide, which they typically cannot penetrate, leading to accumulation. Charge is then balanced by accumulation of electrons on the oxide side of the interface, forming an electric double layer (EDL). The primary advantage of the EDLT is that, in a simple picture, the EDL is a parallel plate capacitor with thickness ~1 Debye length, generating very large capacitances, up to 100 μF/cm2 [32,33]. These translate to electrostatically accumulated carrier densities up to 1014–1015 cm−2 at Vg within the electrochemical stability window of typical ionic liquids (a few volts), one to two orders-of-magnitude beyond conventional FETs [32]. These densities are large enough, in fact, to induce electronic phase transitions [32]. In oxides alone this has enabled electrical control over zero temperature (T) insulator-metal transitions (IMTs) [32,35], T-dependent IMTs [32,36,37], Mott IMTs [32,36], superconductivity [32,38–40], and magnetic order and properties [41–44].

In 2017, Fujiwara et al. [45] applied ionic liquid gating to nominally undoped PLD-grown epitaxial BSO films and achieved n up to 2 × 1014 cm−2 at Vg = +6 V. This was sufficient to induce an IMT in films thicker than 100 nm, and substantially raise mobility, which was found to scale as µ ∼ n2/3 before decreasing at the highest n [45]. Mobilities of 220 and 300 cm2 V−1 s−1 were obtained at 150 and 50 K, respectively [45], comparable to the best reported in BSO films. While such results are highly encouraging, they also raise questions. For example, data were not obtained by Fujiwara et al. at room temperature, above the freezing point of the ionic liquid, due to high gate current (Ig). Metallicity could also only be induced at high BSO thickness (above ∼105 nm), and a H2O etch was required to generate sufficiently smooth surfaces. Moreover, gate biases of +6 V are very large for ionic liquid EDLTs, and reversibility after bias removal was not reported. The latter point is important, as it is now understood that electrolyte gate response in EDLTs can be not only electrostatic, but also electrochemical [46]. In oxides, V0 can be created by the EDL electric field, for example, resulting in Vg-controlled electrochemical doping, as detected from transport irreversibility [47,48], studies in O2 vs inert atmospheres [47,49–51], and operando probes such as x-ray diffraction [52–54] and absorption spectroscopy [54,55]. V0-based electrochemical response has been thus detected in many oxides, including VO2 [49], TiO2 [51], SrTiO3 [50], SrRuO3 [43], NdNiO4 [54,56], La0.5Sr0.5CoO3−δ [47,53], and YBa2Cu3O7−δ [55]. H+ ions created by hydrolysis of H2O contaminants can also be electrochemically doped into oxides in EDLTs, as in VO3 [57,58], SrCoO2.5 [41], and VO2 [59,60]. Although in their infancy, efforts are underway to understand when and why electrochemical vs electrostatic response occurs in oxide EDLTs [46]. Such mechanistic understanding is lacking for BSO, meaning that full appreciation of the potential and limitations of BSO EDLTs has not yet been achieved.

Here we report on a detailed study of ion-gel-gated, smooth, epitaxial BSO films grown by high pressure oxygen sputter deposition. Expanding substantially on prior BSO EDLT work, a range of initial chemical doping is explored (from n ≈ 1019 to 1020 cm−3, achieved via La and V0 doping), in films of thickness d = 10–50 nm. In contrast to many oxides, largely reversible modulation of transport is found over an exceptionally large Vg range (approaching ±4 V), even at 300 K, in vacuo. Operando synchrotron x-ray diffraction further confirms that no detectable structural changes occur vs Vg. This remarkable reversibility is ascribed to a special situation in BSO: the vacancy formation enthalpy is low, meaning that V0 can form, but their diffusivity is also extremely low, rendering them essentially immobile. Any electrochemical gate response due to V0 is therefore confined to the extreme surface of BSO, rendering it practically indistinguishable from ideal, reversible, electrostatic response. Wide-range modulation of transport is thus explored vs initial doping level, in both accumulation and depletion modes, spanning strongly localized to metallic regimes. In particular, a crossover from variable range hopping to weak localization is electrically tuned, tracked via resistance and magnetoresistance. The important case of electron accumulation at positive Vg is then analyzed in detail, using two-channel conduction analysis with self-consistent Schrödinger-Poisson and Thomas-Fermi modeling to extract accumulation layer electron densities and mobilities. Induced electron densities approaching 1014 cm−2 are shown to improve room temperature mobility by up to a factor of ∼24, reaching 51.4 cm2 V−1 s−1 in sputtered films. These results lay the groundwork for future studies of n-dependent transport in high mobility BSO films, also shedding new light on oxide electrolyte gating mechanisms.

II. EXPERIMENTAL METHODS

Conditions for high pressure oxygen sputter deposition of undoped BSO were reported in our prior work [11]. Briefly, rf sputtering at 65–75 W from a nominally stoichiometric
2 in. ceramic BSO target was performed in 1.5 Torr of ultrahigh-purity O₂, on to a 750 °C substrate [11]. As-grown films were then vacuum annealed at 900 °C for 4 h at <10⁻⁷ Torr to controllably V₀ dope [11,20]. The resultant room temperature n increases with d, from ∼1 × 10¹⁹ cm⁻³ at 10 nm to ∼5 × 10¹⁹ cm⁻³ at 50 nm. This is ascribed to increased densities of compensating defects near the substrate interface, and to possible surface/interface depletion effects [11,20]. The resultant room temperature μ ranges from 0.5 to 20 cm² V⁻¹ s⁻¹. To explore a wider range of initial chemical doping, which was not studied in prior BSO EDLT work [45], 50-nm-thick La-doped BSO films were also grown, via dc high pressure oxygen sputtering. A nominally stoichiometric 2 in. ceramic La₀.₀₂Ba₀.₉₈SnO₃ target was dc sputtered at ∼45 W in 1.9 Torr of ultrahigh-purity O₂, onto 850 °C substrates. The La-doped BSO films used here have n ≈ 1 × 10²⁰ cm⁻³ and μ ≈ 30 cm² V⁻¹ s⁻¹ at 300 K. Note that in all cases in this work, LaAlO₃(001) substrates were used. While this generates large mismatch (−8.6%) it eliminates the possibility of annealing- or gate-induced V₀ in the substrate (LaAlO₃ does not readily form VO), removing a source of erroneous results. All films are thus relaxed. As will be reported in a future publication [61], this choice of substrate has surprisingly little impact on μ. Detailed structural and chemical characterization of high pressure oxygen sputtered BSO was provided previously [11]. For illustration, we report here high-resolution wide-angle x-ray diffraction (WAXRD) and grazing-incidence x-ray reflectivity (GIXR) data, acquired on a Panalytical X’Pert using Cu Kα radiation. Contact mode atomic force microscopy (AFM) data from a Bruker Nanoscope V Multimode 8 system are also shown.

EDLT fabrication employed similar methods to our prior work on other oxides [42,47]. As shown in the top- and side-view schematics in Figs. 1(a) and 1(b), a 1 × 1 mm² BSO channels were defined by Ar ion milling, with a steel mask. Ti(20 nm)/Au(60 nm) gate and channel electrodes were then sputtered using a second steel mask, and a four-wire van der Pauw geometry was employed for channel resistance measurement. Note that Ti-induced reduction of the BSO may occur under the contact region, but that this will only serve to improve ohmicity and lower the contact resistance. “Cut and stick” ion gels [34] consisting of 80 wt. % ionic liquid [1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide ([EMI][TFSI])] in 20 wt. % polymer [poly(vinylidene fluoride-co-hexafluoropropylene) (P(VDF-HFP))] were then applied to form EDLTs. As shown in Fig. 1(a), three sections of gel were applied, one covering the BSO channel, the others ensuring high areal coverage of the gate electrodes. Immediately after application of the gel, devices were transferred to a Quantum Design Inc. Physical Property Measurement System (PPMS) for T-dependent transport measurements from 2–300 K in magnetic fields to 9 T, or to a closed-cycle refrigerator where 300 K Vₕ response was measured in vacuum (<10⁻⁴ Torr). dc excitation was employed to measure channel resistance, using a Keithley 220 current source and Keithley 2002 voltmeter. Vₕ and Iₕ were applied and measured using a Keithley 2400 source-measure unit. Following work on electrolyte-gated La₀.₅Sr₀.₅CoO₃₋₅ [53], operando structural characterization under Vₕ was also employed to measure channel resistance, using a Keithley 220 current source and Keitheley 2002 voltmeter. dc excitation was employed to measure channel resistance, using a Keithley 220 current source and Keithley 2002 voltmeter. Vₕ and Iₕ were applied and measured using a Keithley 2400 source-measure unit. Following work on electrolyte-gated La₀.₅Sr₀.₅CoO₃₋₅ [53], operando structural characterization under Vₕ was also performed.

FIG. 1. Device schematics; structural characterization of BaSnO₃₋₄ epitaxial films. Top view (a) and side view (b) schematics of BaSnO₃ electric double layer transistors. Shown are the 5 × 5 mm² LaAlO₃(001) substrate (white), 1 × 1 mm² BaSnO₃ (BSO) channel (red), Ti/Au electrodes (gold color), and the ion gel (pale blue). Electrodes 1–4 are for van der Pauw measurement, while the others are gate electrodes. I, V, and Vₕ are the injected current, measured voltage, and gate voltage, respectively. Orange and blue circles depict gel cations and anions, while the +/- depict holes/electrons. (c) High-resolution wide angle x-ray diffraction (around the 002 film and substrate reflections) from a LaAlO₃(001)/BaSnO₃₋₄ film of thickness d = 13 nm. Inset: Grazing-incidence x-ray reflectivity from the same film. (d) Contact mode atomic force microscopy image of the same film, revealing root-mean-square roughness σRMS = 1.5 Å.
performed, via synchrotron x-ray diffraction (SXRD) on the 33-ID beamline of the Advanced Photon Source, Argonne National Laboratory, with 20 keV ($\lambda = 0.62$ Å) radiation. Those measurements were made in vacuum ($< 10^{-5}$ Torr), with each $V_g$ applied at 300 K for ~30 min, and SXRD scans being taken in the last ~6 min. As discussed in Supplemental Material Section A (Fig. S1) [62], at $V_g = 0$, at least ~8 scans (i.e., ~50 min of exposure) were possible before detectable beam damage, sufficient to obtain the data presented here.

III. RESULTS AND DISCUSSION

A. Structural characterization

As noted above, detailed structural and chemical characterization of high pressure oxygen sputtered BSO was provided earlier [11]. Illustrative of the films studied here, Figure 1(c) shows high-resolution specular WAXRD from a $d = 13$ nm LaAlO$_3$(001)/BaSnO$_{3-\delta}$ film. Only the 002 film and substrate peaks appear, as expected, in-plane grazing incidence diffraction confirming cube-on-cube epitaxy [11]. The out-of-plane lattice parameter from Fig. 1(c) is 4.123 Å, consistent with our prior work, with the expansion over bulk ascribed to defects rather than residual strain [11]. Finite size fringes around the BSO 002 peak indicate low surface roughness, as does the observation of Kiessig fringes to $\sim 5^\circ$ in GIXR [inset to Fig. 1(c)]. This is corroborated by contact-mode AFM images of the same film [Fig. 1(d)], which show root-mean-square roughness of 1.5 Å over $2.5 \times 2.5 \mu m^2$, along with terraces between unit-cell-high steps. Some evidence of decoration of the surface with particles is seen, which could result from surface contamination or BaO [45,63].

B. Electrolyte gating mechanism

As in prior work on ion-gel-gated La$_{0.5}$Sr$_{0.5}$CoO$_{3-\delta}$ [47], in vacuo response of the channel sheet resistance ($R$) to application and removal of $V_g$ was used to assess reversibility and elucidate mechanisms. Figures 2(c) and 2(d) show example data for a $d = 50$ nm La-doped BSO film, measured at $< 10^{-4}$ Torr and 300 K. This film had initial $n = 8.0 \times 10^{19}$ cm$^{-3}$ and $\mu = 29.9$ cm$^2$ V$^{-1}$s$^{-1}$. Gate voltages of increasing magnitude (up to $\pm 4$ V in 0.5 V steps) were applied and removed on a 20 min cycle, with both negative [Fig. 2(a)] and positive [Fig. 2(b)] polarities. Resultant $R(t)$ traces ($t =$ time) for negative and positive $V_g$ are shown in Figs. 2(c) and 2(d), respectively, normalized to $R_0$, the $t = 0$ value of $R$.

Beginning at negative $V_g$, Fig. 2(c) first shows that $R$ remains stable at $R_0$ for the first 20 min, during which $V_g = 0$. In contrast to electrolyte-gated La$_{0.5}$Sr$_{0.5}$CoO$_{3-\delta}$ [47], and other oxides [40], transport in these BSO EDLTs is thus stable after application of the ion gel, even at 300 K. In La$_{0.5}$Sr$_{0.5}$CoO$_{3-\delta}$ this is only possible below $\sim 260$ K; above this $R$ drifts upward with $t$, ascribed to electrochemical
reduction of the film surface even at $V_g = 0$ [47]. This is a first indication of the relative electrochemical stability of BSO EDLs. Application of $V_g = -0.5$ V in Fig. 2(c) then induces a sharp increase in $R$, followed by a gradual increase over 20 min, reaching $R_{off}/R_0$ of 1.034. The resistance change is positive, consistent with electrostatic depletion of electrons in this initially chemically n-doped film. When $V_g$ is returned to zero, $R$ decreases quickly initially, then more slowly, reaching $R_{off}/R_0$ of 1.006 after 20 min. This is close to 1, i.e., almost fully reversible. The remainder of Fig. 2(c) illustrates how $R_{on}$ and $R_{off}$ evolve at larger magnitudes of negative $V_g$, out to $-4$ V. While long time scale effects are present, and $R_{off}/R_0$ begins to dip below 1 at the highest $|V_g|$, the gating is remarkably reversible. Note that a significant contributor to the relatively sluggish $R(t)$ is the side-gate geometry [Figs. 1(a) and 1(b), used here to enable operando SXRD], which separates the channel and gates by nm distances [47,53]. Figure 2(d) reveals similar characteristics for $V_g > 0$. Long time scale effects are visible, but with only modest irreversibility (i.e., small deviations from $R_{off}/R_0 = 1$), even at large $V_g$. The sign is also as expected, the decrease in $R$ being consistent with electron accumulation.

The data of Figs. 2(c) and 2(d) are summarized in Fig. 2(e) by plotting the $300$ K $V_g$ dependence of the gate effect (defined as $R_{on}/R_0$) and irreversibility (defined as $R_{off}/R_0$), on log$_{10}$ scales. With the latter definition, $R_{off}/R_0 = 1$ corresponds to ideally reversible behavior. For a broader view, Fig. 2(e) shows these ratios not only for the $d = 50$ nm La-doped BSO film from Figs. 2(c) and 2(d), but also three additional $V_{O\text{-doped}}$ BSO films, with $d = 10$, 11, and 13 nm. Initial $300$ K $n$ and $\mu$ values thus span from $1.7 \times 10^{19}$ cm$^{-3}$ and $0.5$ cm$^2$ V$^{-1}$ s$^{-1}$ ($d = 10$ nm, VO$_{doped}$) to $8.0 \times 10^{19}$ cm$^{-3}$ and $29.9$ cm$^2$ V$^{-1}$ s$^{-1}$ ($d = 50$ nm, La doped). Considering the gate effect first (solid symbols, left axis), the polarities of response are as expected in each film, $R_{on}/R_0$ going above 1 at negative $V_g$ (depletion), and below 1 at positive $V_g$ (accumulation). A stronger gate effect is also observed on going from the 50-nm-thick La-doped film to the 13-nm-thick VO$_{doped}$ film, to the 10-nm-thick VO$_{doped}$ film, consistent with the lower initial $n$ and $d$. The gate effect at $d = 11$ nm in fact reaches almost four orders-of-magnitude between $-2$ and $+4$ V, achieved here at 300 K. For the $d = 10$ nm VO$_{doped}$ film, the initial $R(300$ K) becomes so large ($>4$ M$\Omega$/sq) that data at $V_g < 0$ (i.e., in depletion) could not be recorded.

In terms of irreversibility (open points, right axis), the main observation is the relatively small deviations from $R_{off}/R_0 = 1$, which are barely visible for the La-doped and $d = 13$ nm VO$_{doped}$ films. Some irreversibility does arise in the 11-nm-thick VO$_{doped}$ film, but at $V_g = +4$ V, where a two order-of-magnitude gate effect occurs, this is only a factor of $\sim 4$. Similarly, at $V_g = -2$ V, where a gate effect of the opposite polarity of two orders-of-magnitude occurs, the irreversibility is negligible. We thus conclude that the gate effects here are predominantly reversible, even at 300 K, and over a $\pm 4$ V range, independent of initial chemical doping. Moreover, the limited irreversibility that does arise at large $V_g$ magnitudes can also be shown to be associated with the ion gel, not the BSO. Working with $d = 11$ nm VO$_{doped}$ films, for example, $V_g$ of $\pm 4$ V was applied for 20 min, $V_g$ returned to zero, the ion gel removed, and the BSO film surface cleaned. The latter was achieved by sonication in acetone, followed by baking in vacuum at 100 °C. As shown in Supplemental Material Section B (Fig. S2) [62], $R$ returns to within 50% of its initial value after this process, and the BSO can be regated with a new ion gel, demonstrating that the film is not irreversibly damaged. We thus ascribe what limited irreversibility occurs in Fig. 2 to electrochemical degradation of the ion gel, rather than the BSO.

The above picture is further supported by Fig. 2(f) which plots the $V_g$ dependence (to $\pm 4$ V, both ascending and descending $V_g$) of $I_g$ for the $d = 11$ nm VO$_{doped}$ BSO film in Fig. 2(e). The data are largely devoid of the peaks that would indicate distinct electrochemical processes [41,64] (except perhaps at $-3.25$ V, see below), and are instead dominated by only gradual onset of significant $|I_g|$ at the highest $|V_g|$. In these devices, with 1 mm$^2$ channels, $|I_g|$ in fact only exceeds 0.1 $\mu$A at $V_g = -3.15$ V and $V_g > 3.04$ V. We ascribe this current to electrochemical degradation at the very highest $V_g$, mostly of the gel, but potentially also involving residual H$_2$O$_x$ [41]; the small potential peak at $-3.25$ V is likely of the same origin. More specifically, electrochemical consumption of cations (anions) in the gel at the BSO surface at positive (negative) $V_g$ would lead to interfacial anion (cation) excess after removal of $V_g$, resulting in $R_{off}$ being larger (smaller) than $R_0$, as observed in Fig. 2(e). Similar phenomena may also explain the small peak in irreversibility around $V_g = 2$ V in Fig. 2(e); hydrolysis of residual H$_2$O could lead to excess hydroxide ions at the BSO surface and thus $R_{off} > R_0$. This $\sim 2$ V irreversibility peak could not be eliminated even by 3 h of in situ SXRD annealing at $10^{-3}$ Torr, potentially due to the known difficulty of removing residual water [41]. As a final comment on Fig. 2(f), we note that the $|I_g|$ recorded here at 300 K are certainly sufficiently low to enable reliable $V_{g-c}$ and $T$-dependent BSO transport studies. We are thus not restricted to low $T$ [45].

Further support for unusual reversibility over an exceptional $V_g$ window is provided by operando SXRD. Figures 3(a) and 3(b) show 300 K specular SXRD data collected around the 002 film and substrate reflections for an ion-gelled $d = 13$ nm VO$_{doped}$ BSO film at $V_g = 0$, $-1$, $-2$, $-3$ V [Fig. 3(a)] and $V_g = 0$, +1, +2, +3, +4 V [Fig. 3(b)]. The $V_g = 0$ BSO 002 peak occurs at $L = 1.839$ [the $L$ axis is in substrate reciprocal lattice units (r.l.u.)], corresponding to an out-of-plane lattice parameter of 4.125 Å, consistent with Fig. 1(c). The striking feature at nonzero $V_g$ is the almost complete absence of changes with gating. Some evidence of a barely detectable decrease in the amplitude of BSO Laue fringes may arise at $V_g = -3$ and $+4$ V, but could also be due to minor beam damage. Aside from this, the BSO Bragg peak position, peak width, and fringe structure are essentially $V_g$ independent. Note here that in situ SXRD measurements were performed on the beamline during SXRD data acquisition (Supplemental Material Section A [62]) confirming that gating did take place. As a point of comparison, gate-induced VO formation in an electrochemically active material such as La$_0.5$Sr$_0.5$CoO$_{3-\delta}$ leads to very large (1%) out-of-plane lattice parameter expansion at $V_g$ of only $+2$ V, accompanied by strong damping of Laue oscillations [53].

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post-growth reduction (as used in this work to V\textsubscript{O} dope) is effective only in thin films, not bulk single crystals [10]. V\textsubscript{O} still occurs in the latter, but their density is locked-in during growth [10]. The result of this situation is that under the gating conditions used here (300 K, 20 min) the BSO V\textsubscript{O} diffusion length, \( l = (D_t)^{1/2} \approx 1 \) Å. Any V\textsubscript{O} that do form under \( V_g \) are therefore confined to the extreme surface region of the BSO. We propose that this special situation is the origin of the strikingly reversible, wide-\( V_g \)-window behavior seen here at 300 K in BSO EDLTs, and also recently in SrSnO\textsubscript{3} EDLT\textsubscript{s} [68]. Electrochemical effects due to V\textsubscript{O} are strongly suppressed by the exceptionally low V\textsubscript{O} diffusivity, meaning that redox response, if present, is essentially indistinguishable from ideal electrostatic accumulation and depletion of electrons. With this insight, future work studying electrochemical vs. electrostatic response in electrolyte-gated oxides with systematically varied \( \Delta H_{V_{o}} \) and \( D_{V_{o}} \) would clearly be worthwhile. With regard to gating mechanisms involving \( H^+ \), liberated by potential hydrolysis from residual H\textsubscript{2}O, we emphasize that the very long time scale effects and large \( R_{in}/R_{0} \) characteristic of electrolyte-induced \( H^+ \) doping [41,57–60] are absent here.

**C. Temperature-dependent transport**

Armed with understanding of the mechanisms of electrolyte gating in BSO EDLTs, we now progress to discussion of \( T \)-dependent transport. Shown in Fig. 4 are \( R(T) \) plots at multiple \( V_g \) [Figs. 4(a), 4(d), and 4(g)], along with the \( V_g \) dependence of the apparent 2D Hall electron density \( n_{H}^{\text{3D}} \) [Figs. 4(c), 4(f), and 4(i)] and mobility \( \mu_{H} \) [Figs. 4(b), 4(e), and 4(h)], at \( T = 2, 150, \) and 300 K. Data are shown for three films: \( d = 50 \) nm La-doped BSO (top panel), \( d = 12 \) nm V\textsubscript{O}-doped BSO (middle panel), and \( d = 10 \) nm V\textsubscript{O}-doped BSO (bottom panel), with systematically decreasing initial 3D Hall density \( (n_{H}^{\text{3D}}) \) and mobility \( (\mu_{H}) \), as given in the figure. It should be emphasized here that \( n_{H}^{\text{3D}}(V_g) \) and \( \mu_{H}(V_g) \) in these plots are apparent values, directly extracted from Hall data, not accounting for current shunting due to the doped BSO region under the accumulation/depletion layer at the interface with the ion gel. The further analysis required for us to determine electrostatic doping depth profiles, and thus more quantitatively understand such data, is provided in Sec. III D.

Starting with the \( d = 50 \) nm La-doped BSO film with \( n_{H}^{\text{3D}} = 8.0 \times 10^{10} \) cm\textsuperscript{-2} and \( \mu_{H} = 29.9 \) cm\textsuperscript{2}V\textsuperscript{-1}S\textsuperscript{-1}, Fig. 4(a) shows typical \( V_g = 0 \) behavior at such relatively heavy initial doping. [Note that \( t \) wo \( V_g = 0 \) curves are shown in Fig. 4(a), for reasons explained in the caption]. \( R \) first decreases on cooling from 300 K (metalliclike behavior) before presenting an upturn below \( \sim 50 \) K; such behavior will be shown below to be due to weak localization. Application of \( V_g = \pm 4 \) V (i.e., electron accumulation/depletion) then results in modulation of low \( T \) resistance by \( \sim 32 \) and \( +39 \%), respectively, the relatively modest changes being due to the high thickness and initial chemical doping. The only change in the form of \( R(T) \) is weakening of the low \( T \) upturn at lower \( R \), i.e., at positive \( V_g \). Weakly localized transport is thus apparent at all \( V_g \) in Fig. 4(a), approaching positive \( dR/dT \) at all \( T \) at the highest accumulated electron densities, where \( R \) is \( \sim 1\% \) of \( h/e^2 \) [i.e., 26 kΩ (\( h \) = Planck’s constant, \( e \) = electronic charge)]. We
FIG. 4. Temperature- and gate-voltage-dependent transport in Ba$_{1-x}$La$_x$SnO$_{3-y}$ films. Temperature ($T$) dependence of the sheet resistance ($R$) at various $V_g$ for (a) 50-nm-thick LaAlO$_3$(001)/
LSCO/2nm BaO$_{9.88}$SnO$_3$, (d) 12-nm-thick LaAlO$_3$(001)/BaSnO$_{3-y}$, and (g) 10-nm-thick LaAlO$_3$(001)/BaSnO$_{3-y}$. (b) and (c), (e) and (f), and (h) and (i) show corresponding $V_g$ dependences of the apparent Hall mobility $\mu_H$ and 2D Hall electron density $n_{2D}$ at 300, 150, and 2 K for the same films. The labels above the panels indicate the film type, thickness ($d$), and initial 3D Hall electron density $n_{2D}$ and Hall mobility $\mu_0$. In (h) and (i), 2 K data could not be collected due to the large resistances. Note that in (a)–(c) two sets of data are shown at $V_g = 0$. In this particular device, positive $V_g$ was applied first, exploring magnitudes as large as 6 V. Some shift in $R$ was thus found on returning to $V_g = 0$ and performing measurements at negative $V_g$. Note parenthetically here that the irregular features in $R(T)$ in the 200–300 K range are associated with the freezing point of the ionic liquid, and are common in EDLTs [35,69]. As shown in Figs. 4(b) and 4(c), $n_{2D}$ and $\mu_H$ evolve qualitatively as expected with $V_g$ and $T$. Specifically, at 300 K, $n_{2D}$ increases by 12% with $V_g$ up to +4 V, consistent with additional electron accumulation, whereas $n_{2D}$ decreases by 10% with $V_g$ down to −4 V, consistent with depletion. Again, the relatively small $V_g$-induced changes in $n_{2D}$ are due to the high thickness and initial chemical doping, which render electrostatic effects modest. The resulting $\mu_H$ increases monotonically from −4 to +4 V, consistent with the typical increase in $\mu$ with $n$ in BSO films [20], due to screening of charged defects. Moving to $T$ dependence, $n_{2D}(T)$ is seen to be weak [Fig. 4(c)], consistent with the expected degenerate doping at these doping levels [11,20]. $\mu_H$, on the other hand, increases by as much as 26% on cooling from 300 to 2 K at $V_g = 0$, indicative of significant electron-phonon scattering at these relatively high $\mu$ [11,20].

The $d = 12$ nm VO$_{0.75}$-doped film (middle panel) with $n_{2D} = 4.1 \times 10^{19}$ cm$^{-3}$ and correspondingly lower $\mu_0 = 8.4$ cm$^2$V$^{-1}$s$^{-1}$ shows significantly different behavior. $R(300 \text{K})$ at $V_g = 0$ is ~30 times higher than in the 50 nm La-doped case, $R(T)$ showing a ~10-fold increase on cooling to 2 K. The initial state is thus significantly more insulating, consistent with the lower $n_{2D}$. Electron depletion at $V_g$ down to −3 V then results in strongly localized transport, shown in Sec. III E to be due to variable-range hopping. Conversely, electron accumulation at $V_g$ up to +4 V results in rather flat $R(T)$ with $R < h/e^2$ (26 kΩ). As detailed in Sec. III E, this device thus presents an apparent strong- to weak-localization crossover, spanning well over four orders-of-magnitude in low $T$ resistance. As shown in Figs. 4(e) and 4(f), the corresponding behaviors of $n_{2D}$ and $\mu_H$ are qualitatively as expected. The gate-induced modulation of $n_{2D}$ is much stronger than in the $d = 50$ nm La-doped BSO device, due to the lower initial doping. $n_{2D}$ in fact becomes unmeasurable at the largest negative $V_g$ and lowest $T$ due to high $R$; related to this, in Fig. 4(f) at low $T$ and −1 V, hopping conduction likely makes $\mu_H$ somewhat unreliable. Consistent with the lower initial doping, even at the highest accumulated electron densities $\mu_H$ reaches only ~18 cm$^2$V$^{-1}$s$^{-1}$, with no increase on cooling, indicative of dominant defect scattering [20].

Finally, the $d = 10$ nm VO$_{0.75}$-doped BSO film, with $n_{2D} = 1.7 \times 10^{19}$ cm$^{-3}$ and $\mu_0 = 0.5$ cm$^2$V$^{-1}$s$^{-1}$ shows yet different behavior. $R(T)$ before ion gel application in this case is already strongly insulating, $R$ exceeding 10$^7$ Ω at ~100 K. This is consistent with the expected high defect density at low $d$ in highly lattice-mismatched BSO, which will generate compensation, trapping, strong scattering, and surface/interface depletion. As noted in the discussion of Fig. 2(e), the electrostatic depletion regime can thus not be studied in such devices due to very high $R$. Application of positive $V_g$ up to 4 V results in $R < 10^7$ Ω, however, accompanied by weak $R(T)$, meaning that a crossover from strongly localized to weakly localized/metallic transport remains possible at low $n_0$. This is consistent with the study of Fujiwara et al. [45] on nominally undoped BSO, although very thick films (100s of nm), and gate biases up to +6 V (potentially beyond the electrochemical stability window of the ionic liquids [33,70]) were required to achieve metallicity in that work. One
other notable feature in Fig. 4(g), found only in such highly insulating films, is the non-negligible effect of ion gel application even at \( V_g = 0 \). At 300 K, \( R \) increases by a factor of \( \sim 6 \) after gel application, likely indicating ion-gel-induced doping even at \( V_g = 0 \). This is not uncommon in EDLTs [32,33,71], and can derive from preferential association of one ion type with the semiconductor surface, amplified here by the strong \( \mu(n) \). Moving on to Figs. 4(h) and 4(i), \( n_{3D}^H \) and \( \mu_H \) are now easily interpreted due to the low thickness and highly insulating starting BSO. Under these conditions the accumulation layer at positive \( V_g \) essentially carries all of the current. The induced electron density reaches \( 6 \times 10^{13} \text{ cm}^{-2} \), within a factor of 2 of Fujiwara et al. [45], and in the typical range for semiconducting EDLTs [32,33]. Significantly, \( \mu_H \) at 300 K is increased from 0.5 to 11.9 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \) (i.e., a factor of 24) by this interfacial electron accumulation, as discussed below. As already noted, however, extracting accurate corresponding \( \mu(n) \) from heavier doped, thicker, films [as in Figs. 4(a)–4(f)] requires knowledge of the depth profile of the accumulated electron density.

**D. Quantitative analysis of electrostatic doping profiles and accumulation layer parameters**

Extraction of estimated accumulation layer electron densities and mobilities from the \( V_g \)-dependent data discussed above was achieved by modeling the depth profile of electrostatic doping and applying a two-channel conduction model. In this manner, transport parameters in the accumulation layer near the interface with the ion gel at positive \( V_g \) can be separated from those of the underlying BSO, even in the presence of initial chemical doping. In the absence of known \( \mu(n) \), models featuring continuous variation of \( n \) and \( \mu \) with depth \( z \) are not feasible [72], and so a discrete two-channel parallel conduction model was applied. In such a model [72,73],

\[
\begin{align*}
\mu_H & = n_{3D}^H \mu_A d_A + n_{3D}^B \mu_B d_B, \\
\mu_H^2 & = n_{3D}^H \mu_A^2 d_A + n_{3D}^B \mu_B^2 d_B,
\end{align*}
\]

where \( n_{3D}^H \) and \( \mu_H \) are the apparent Hall electron density and mobility (as shown in Fig. 4), \( n_{3D}^A \) and \( \mu_A \) are the 3D carrier density and mobility in the accumulation layer, and \( n_{3D}^B \) and \( \mu_B \) are the 3D carrier density and mobility in the “bulk” region, under the accumulation layer. This bulk region is considered to have identical properties to starting films, i.e., \( n_{3D}^B = n_0 \) and \( \mu_B = \mu_0 \). In addition, the thicknesses of the accumulation and bulk layers obey \( d_A + d_B = d \). Extraction of \( n_{3D}^A \) and \( \mu_A \) from the data shown in Fig. 4 (as well as \( n_{3D}^A \)) then only requires estimation of \( d_A \), which we define as the thickness of the region in which 90% of the electrostatically induced electrons are confined.

To fully understand the induced carrier depth profile, and accurately determine \( d_A \), both Thomas-Fermi (TF) approximation [74] and self-consistent Schrödinger-Poisson (SP) numerical modeling [75] was performed. A full description is provided in Supplemental Material Section C (including Fig. S3) [62]. Briefly, the TF approximation treats electrons in the accumulation layer as a 3D free electron gas, which is appropriate when \( k_T d_A \gg 1 \), where \( k_T = (3 \pi^2 n_{3D}^A)^{1/3} \) is the Fermi wave vector [74]. TF calculations were performed by solving the Poisson equation,

\[
\frac{d^2 \phi(z)}{dz^2} = -\frac{4 \pi \rho_e(z)}{\varepsilon_r},
\]

where \( \phi(z) \) is the potential profile, \( \rho_e(z) \) is the charge density profile, and \( \varepsilon_r \) is the relative permittivity. In the TF approximation [74] this is supplemented with the relation

\[
e\phi(z) = (3\pi^2)^{2/3} \frac{\hbar^2}{2m_e} [n_{3D}(z)]^{2/3},
\]

where \( m_e^* \) is the electron effective mass. Combining Eqs. (3) and (4) with standard boundary conditions then enables numerical computation of \( n_{3D}(z) \) and \( \phi(z) \). More accurate SP modeling, on the other hand, treats the accumulation layer electrons as a 2D quantum mechanical gas confined in the potential well created by \( V_g \) at the BSO/ion gel interface. This is done by self-consistently solving Eq. (3) with the Schrödinger equation. For a 2D electron gas in the effective mass approximation this reduces to solving a 1D Schrödinger equation for an envelope function \( \psi(z) \) [75],

\[
-\frac{\hbar^2}{2m_e^*} \frac{d^2 \psi(z)}{dz^2} + [-e\phi(z) - E] \psi(z) = 0.
\]

The charge density \( \rho_e(z) \) is then expressed as a sum of the electron density in each band:

\[
\rho_e(z) = e \left\{ n^+_g - \beta k_B T \sum_i |\psi_i(z)|^2 \right\} \times \ln \left[ 1 + \exp \left( -\frac{E_i - E_F}{k_B T} \right) \right],
\]

where \( n^+_g \) is the density of ionized donors, \( E_F \) is the Fermi energy, \( \beta = \frac{m_e^*}{\hbar^2} \), \( k_B \) is Boltzmann’s constant, and \( \psi_i \) and \( E_i \) are the normalized envelope wave function and eigenenergy of the \( i \)-th state [75]. Self-consistent calculations then proceed by numerically solving (5) using the TF method as an initial input, then inputting the result into (6). The Poisson equation (3) is then solved and the process repeated until convergence is achieved. Details are provided in Supplemental Material Section C [62]. Note that while the SP method is implemented here at 300 K, and TF is a \( T = 0 \) theory, for the degenerately doped films studied here the SP results are very similar at 0 and 300 K.

\( V_g \)-dependent electrostatically-induced electron distributions \( [n_{3D}(z)] \) calculated via these TF (blue curves) and SP methods (red curves) are shown in Fig. 5(a) for an illustrative \( d = 50 \text{ nm} \) La-doped BSO film. Here \( z = 0 \) is placed at the substrate/BSO interface, so that \( z = 50 \text{ nm} \) corresponds to the BSO/ion gel interface. The qualitative features in Fig. 5(a) are essentially as expected. The two approaches agree in the tail of the \( n_{3D}(z) \) distribution (approaching \( 8 \times 10^{19} \text{ cm}^{-3} \), the initial chemical doping level), but differ at the BSO/ion gel interface. In the latter limit the TF approach yields a \( V_g \)-dependent constant at \( z = 50 \text{ nm} \), inherent to a boundary condition in the model (see Supplemental Material Section C [62]), whereas the SP calculations yield \( n_{3D} \to 0 \) as the ion gel is entered. A peak in \( n_{3D}(z) \) thus forms in the SP data, in this case at \( \sim 1 \text{ nm} \) from the interface, but of course shifting towards...
La$_{0.02}$Ba$_{0.98}$SnO$_3$, $d = 50$ nm, $T = 300$ K

(a) 3D charge carrier density $n^{3D}$ vs depth $z$ in 50-nm-thick LaAlO$_3$(001)/La$_{0.02}$Ba$_{0.98}$SnO$_3$ at various $V_g$, calculated using the Thomas-Fermi (TF) approach (blue shades) and the self-consistent Schrodinger-Poisson (SP) approach (red shades). The La$_{0.02}$Ba$_{0.98}$SnO$_3$/ion gel interface is at $z = 50$ nm. The red shaded area indicates 90% of the induced electron density at $V_g = 4$ V, which is confined within 4 nm (vertical red dotted line) of the interface. The horizontal red dashed line marks $n^{3D}$ at $V_g = 0$. Inset: Corresponding electrostatic potential vs $z$ and (c) $V_g$ dependence of the induced 2D electron density $\Delta n^{2D}$ and accumulation layer mobility $\mu_A$ extracted via the SP approach for the three devices in Fig. 4 (50-nm-thick LaAlO$_3$(001)/La$_{0.02}$Ba$_{0.98}$SnO$_3$, 12-nm-thick LaAlO$_3$(001)/BaSnO$_3$ and 10-nm-thick LaAlO$_3$(001)/BaSnO$_3$ films).

the interface with increasing $V_g$ [76]. Importantly, however, with both methods, $\sim 90\%$ of the electrostatically induced electrons reside in a $\sim 4$-nm-thick channel (vertical red dotted line) at the BSO/ion gel interface, only weakly dependent on $V_g$. Accurate values for $d_A$ can thus be confidently determined for use in Eqs. (1) and (2). We note that for $d_A \approx 4$ nm, the condition for validity of the TF approximation can be checked. It can be seen that $k_Fd_A > 1$ when $n_A^{3D} \geq 5.3 \times 10^{13}$ cm$^{-3}$; the TF approximation is thus applicable for all BSO films in this work, where $n_A^{3D} \geq 1.7 \times 10^{13}$ cm$^{-3}$. For completeness, shown in the inset to Fig. 5(a) are the accompanying $V_g$-dependent potential profiles $\phi(z)$, which are set to zero deep in the film. Similar shapes are obtained from the TF and SP methods, but with generally lower magnitudes than those predicted by simple modeling. This is because these models ignore significant factors such as traps, localized states, and electrochemistry, and rely on the effective mass approximation and so on. We note that the TF method, despite its simplicity, is able to capture the main features of the electron density and potential profile, providing a good estimate of $d_A$ and $\phi(z)$.

Calculations of this type were repeated for all films in this study, to determine $d_A$ values. Equations (1) and (2) were then applied to the data shown in Fig. 4 to extract $n_A^{3D}$ (and thus $n_A^{3D}$) and $\mu_A$. Figures 5(b) and 5(c) summarize the results by plotting the 300 K accumulation-mode $V_g$ dependence of the induced 2D electron density, $\Delta n^{2D}$ [Fig. 5(b)] and $\mu_A$ [Fig. 5(c)]. $\Delta n^{2D}$ is similar for all three films shown, reaching almost $8 \times 10^{13}$ cm$^{-2}$ at $V_g = +4$ V. This is similar to reported values in many ionic-liquid- or ion-gel-gated inorganic and organic semiconductors [32,33], and within a factor of $\sim 2$ of the Fujiwara et al. BSO EDLT result [45]. The latter was obtained at $+6$ V, however, and in our case we achieved this with demonstrated reversibility. Figure 5(b) also shows slightly reduced $\Delta n^{2D}$ as the initial chemical doping $n_A^{3D}$ is decreased (e.g., in the $d = 10$ nm VO-doped case), which we ascribe to a lower accessible density of extended states, and higher trap density.

The corresponding $\mu_A(V_g)$ behavior in Fig. 5(c) reveals substantial increases in mobility with electron accumulation in all cases. The $d = 50$ nm La-doped BSO device with initial mobility of 29.9 cm$^2$ V$^{-1}$ s$^{-1}$ is increased to 51.4 cm$^2$ V$^{-1}$ s$^{-1}$ (a 72% increase), while the $d = 12$ nm VO-doped BSO with initial mobility 8.4 cm$^2$ V$^{-1}$ s$^{-1}$ is increased to 18.4 cm$^2$ V$^{-1}$ s$^{-1}$ (a 120% increase). This mobility boost is consistent with the general increase in $\mu$ with $n$ seen in BSO, due to improved screening of charged defects, particularly dislocations [1,2,4,13,20]. Ion gel gating enables this without increasing the ionized impurity density, however. A future study is planned to examine $\mu_A(n)$ in this manner in even higher quality MBE-grown films with larger $\mu_0$ [77]. We note for now, however, that the as-deposited La-doped BSO and peak gated mobilities reported here (29.9 and 51.4 cm$^2$ V$^{-1}$ s$^{-1}$, respectively), are outstanding for sputtered BSO. The factor of 24 improvement from 0.5 to 11.9 cm$^2$ V$^{-1}$ s$^{-1}$ in the $d = 10$ nm BSO EDLT is also noteworthy. The initial mobility in such films is suppressed by strong disorder and compensation due to the low $d$ (particularly due to misfit dislocations...
As discussed in the next section, such mobility increases are very likely driven by strong to weak localization crossovers, i.e., from hopping conduction to weakly localized hopping, i.e., from hopping conduction to weak localization. The solid lines in (d) are fits to a 2D weak localization model yielding Thouless lengths $l_T$ of 80–140 nm, as described in the text. The $V_g$ legend at the top applies to all panels.

3D Mott variable-range hopping (Mott VRH), for example [79–81].

As shown in Fig. 6(a), this initial BSO film, and all negative $V_g$ curves (i.e., electron depletion), present approximately straight lines with negative slope in the low $T$ limit on this Zabrodskii plot, indicating some form of exponential $T$ dependence. As indicated by the black solid lines, $m = 0.5$ is supported from 0 to $-3$ V, setting in at some $V_g$-dependent $T$ between $\sim 70$ and 10 K. The widest ranges of adherence to $m = 0.5$ (at $V_g = -1$, $-2$ V) reach one order-of-magnitude in $T$ and almost two orders-of-magnitude in $\rho$. Extracted $T_0^{\text{ES}}$ values range from 370 to 40 K, and the $T$-dependent VRH activation energies safely exceed the lowest measurement temperature at $V_g < -1$ V. We thus conclude that ES VRH is active at low $T$ between 0 and $-3$ V [82]. As is often the case, however, ES VRH gives way to a weaker $T$ dependence at higher $T$ [79,83,84], in this case with $m = 0.25$ [red solid lines in Fig. 6(a)]. This indicates a crossover to Mott VRH [79,83,84]. This is supported by analysis of the scaling between $T_0^{\text{ES}}$ and $T_0^{\text{Mott}}$, which follows the expected relation, as shown in Supplemental Material Section D, Fig. S4 [62]. As also shown in Fig. 6(a), positive $V_g$ (i.e., electron accumulation) results in very different behavior: In $W$ becomes negative, the slope of ln $W$ vs ln $T$ becoming small before inverting to positive slope at the highest $V_g$. This indicates a reduced activation energy that decreases on cooling, i.e., nonexponential, metalliclike behavior [78].

To further clarify the origin of this crossover from exponential (ES VRH) to logarithmic $T$ dependence, low $T$ perpendicular field magnetoresistance (MR) data were taken and analyzed. $MR = [(\rho(H_\perp) - \rho(0))/\rho(0)] \times 100\%$ is plotted vs perpendicular (out-of-plane) field $H_\perp$ in Fig. 6(c), at 2 K, for all $V_g$ at which 2 K measurements were possible (−1 to +4 V). At $-1$ V, where Fig. 6(a) shows positive $W$ at low $T$ and clear ES VRH, MR($H_\perp$) is clearly dominated by a positive component. As shown by the blue solid line, the positive MR component is of the form

$$\ln \left( \frac{R}{R(H_{\perp} = 0)} \right) = s e^2 \frac{L^3}{h^2} \frac{H_{\perp}^2}{T} \frac{T_0^{\text{ES}}}{T}^{3/2},$$

where $s = 0.0015$, and $L_e$ is the electron localization length. This is as expected for ES VRH, where perpendicular fields reduce wave function overlap, generating positive MR [79], the deviations at high field being ascribable to factors such as other MR contributions, weak-to-strong-field limits, minor self-heating, etc. The extracted localization length in this case is 11 nm. The negative MR component at low field then grows with electron accumulation, $V_g$ from 0 to $+4$ V resulting in sign reversal to entirely negative MR, characteristic of the field-induced dephasing effect in weak localization [79,81].

Quantitatively confirming this, Fig. 6(d) shows the 0 to $+4$ V MR data replotted as $-\Delta R/R^2$ vs $H_{\perp}$, along with fits (solid...


\[ \frac{-\Delta R}{R^2} = \frac{e^2}{2\pi^2 \hbar} \left[ \Psi \left( \frac{1}{2} + \frac{h/2e}{l_{el} H_0} \right) - \ln \left( \frac{h/2e}{l_{el} H_0} \right) \right], \quad (9) \]

where \( \Psi \) is the digamma function, and \( l_{el} \) and \( l_{in} \) are the elastic and inelastic scattering lengths \([79,81]\). This expression for weak-localization-driven negative perpendicular field MR (i.e., positive magnetoconductance) is seen to describe the data well, yielding Thouless lengths \( l_{Th} = \frac{\sqrt{e^2}}{\hbar} \) that increase from 80 to 140 nm with electron accumulation. Importantly, in all cases \( l_{Th} \gg d \), consistent with 2D weak localization. The analysis in Fig. 6 thus provides compelling evidence of a gate-tuned strong to weak localization crossover, highlighting the utility of electrolyte gating for wide-range modulation of the electronic properties and ground state in BSO.

### IV. SUMMARY

In conclusion, ion gel gating of thoroughly characterized epitaxial BaSnO\(_3\) thin films has been shown to enable wide-range modulation of electron density, resistance, and mobility, studied vs film thickness and initial chemical doping level. Gate-voltage-dependent measurements of \textit{in vacuo} irreversibility, gate current, and structure (from \textit{operando} synchrotron x-ray diffraction) reveal a remarkable level of reversibility, even at 300 K. We argue that this occurs due to a special situation in BaSnO\(_3\), where oxygen vacancy diffusion is so slow that electrochemical electrolyte gate response is strongly suppressed, resulting in behavior indistinguishable from simple electrostatic accumulation and depletion of electrons. Calculations of depth profiles of electrostatic doping were then used in conjunction with two-channel conduction modeling to extract accumulation layer electron densities and mobilities, even in the presence of initial chemical doping. Strong enhancement of mobility with electron accumulation is deduced, reaching a factor of 24 and resulting in peak room temperature mobility of 51.4 cm\(^2\) V\(^{-1}\) s\(^{-1}\) in sputtered LaAlO\(_3\)/La\(_{0.02}\)Ba\(_{0.98}\)SnO\(_3\) (50 nm) films. Finally, a gate-tuned crossover from strong to weak localization has been established, verified by detailed analysis of temperature- and magnetic-field-dependent transport. These results shed important new light on oxide electrochemical gating mechanisms in general, in addition to paving the way for future electron-density-dependent studies of high mobility BaSnO\(_3\) thin films.

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[82] There is some evidence in Fig. 6(a) and Fig. S4 of deviations from ES VRH towards some strong $T$ dependence (i.e., $m > 0.5$) at low $T$. Low $T$ studies are required, but there are known effects capable of this, e.g., I. Terry, T. Penney, S. von Molnár, and P. Becla, Phys. Rev. Lett. **69**, 1800 (1992).
